Electronic Supporting Information for:

Large scale boron carbon nitride nanosheets with enhanced lithium

storage capabilities

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Experimental Section

Material Synthesis: LiCl and KCl (Aldrich, 45/55 wt) were mixed and ground in a mortar. The resulting mixture LiCl/KCl was evacuated at 200 °C for 4 days and transferred into an argon-filled glovebox. Sodium borohydride (2 mmol) and urea (1 mmol) were added into the eutectic mixture LiCl/KCl (2.5 g), and then the powder mixture was ground with a Retsch P7 ballmill for 2 min. The resulting powder was heated to 750 °C at a rate of 10 °C•min⁻¹ for 2 h under nitrogen flow (5L min⁻¹). After being cooled to room temperature, the obtained powder was washed with deionized water, recovered by filtration, washed twice with water and then dried under vacuum at room temperature overnight. Finally, 200 mg BCN sample were obtained.

Material Characterization: XRD patterns were collected on a Panalytical X'Pert PRO diffraction system using Cu Kα radiation. SEM was performed on a Zeiss Supra 55 VP instrument. TEM was performed on a JEOL 2100 (operating at 110kV). HRTEM was carried out on a JEOL JEM-2200FS instrument working at 200 kV. AFM measurements were performed on a cypher atomic force microscope. XPS was performed using a Thermo Scientific K-Alpha ESCA instrument equipped with Al Kα monochromatized radiation at 1486.6 eV X-ray source. Photoelectrons were collected from a take-off angle of 90° relatively to the sample surface. Charge referencing was done by setting the lower binding energy C1s photopeak at 285.0 eV C1s hydrocarbon peak. Nitrogen sorption isotherms were obtained using a Quadrasorb apparatus at 78 K. The specific surface area was calculated by the Brunauer-Emmett-Teller (BET) method.

Electrochemical measurements: An electrode slurry was prepared using 70 wt% active material (fewlayer BCN nanosheets), 15 wt% acetylene black (Super P, Timcal), and 15 wt% polyvinylidene fluoride (PVDF, Aldrich) binder dissolved in N-methyl-2-pyrrolidone (NMP, Aldrich). The slurry was coated on Cu foil covering an area around 1 cm² and dried at 100 °C in vacuum overnight to remove the solvent before pressing. The material loading on the Cu foil was approximately 1 mg cm⁻². Coin cells (CR2032) were fabricated using Li metal as the counter electrode, and 1M LiPF₆ in 1:1 w/w ethylene carbonate:diethyl carbonate as electrolyte. The cells were assembled in an argon-filled glovebox. Galvanostatic cycling tests of the assembled cells were carried out on a Solartron 1470E electrochemical workstation in the voltage range of 0.01–3.0 V (vs. Li⁺/Li). The cycled cell was disassembled and the electrode was washed by dimethyl carbonate in the glove box and analysed by XRD.



Fig. S1 XRD pattern of the sample.



Fig. S2 Nitrogen adsorption and desorption isotherm (a) and BJH pore size distribution of BCN nanosheets (b).



Fig. S3 XPS spectra of BCN nanosheets: (a) B1s, (b) C1s, and (c) N1s. The chemical states of B, C, and N elements were further investigated using X-ray spectroscopy (XPS, details in SI) at the B1s, C1s, and N1s core levels. The B1s spectrum highlights a dominating component centered at 190.6 eV, accompanied by a small component peaked at 191.8 eV. The dominant peak is attributable to B—N bonds, while the high energy component is assigned to B—O bonds.¹⁶ The C1s shows three components at 285.0, 286.5, and 288.9 eV, corresponding respectively to C—C, C=N or C≡N, and C—N bonds.^{7,16} In the N1s spectra, the peak at 398.1 eV is assigned to B—N bonds.^{7,16} A shoulder at higher energy (*ca.* 399 eV) accounts for the formation of C—N or C≡N bonds.^{7,16}



Fig. S4 Enlarged XRD patterns of BCN nanosheets electrode materials before and after cycling for 5000 times at $2 \text{ A} \cdot \text{g}^{-1}$, showing excellent structure durability of BCN nanosheets.



Fig. S5 FTIR spectra of BCN nanosheets.